

Volumetric Method of Moments (V-MoM) in Modelling Nanotopologies: A Review

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Abstract—This paper provides a summary review of our group's work on the modeling of nanoantennas. By utilizing a Volumetric Method of Moments (V-MoM) algorithm, we solve the light-nanoantenna problem on three different levels: I) the most general solution to Maxwell's equations, i.e. the linear response of a nanoantenna to a given incident light and a specific frequency; II) an incident light independent solution, i.e. the frequency dependent eigenmodes of a nanoantenna and III) an solution that is both excitation and frequency independent, i.e. the natural mode of a nanoantenna. Through such a treatment, a set of tools that can systematically treat the interaction of light with a nanoantenna is developed and is hoped to pave the road for the future nanoantenna design.

Index Terms—Nanoantennas, Volumetric Method of Moments (V-MoM), Eigenmode Expansion Method (EEM), Natural Frequency Analysis.

I. INTRODUCTION

In the last decade, thanks to the grand leap in nanotechnology, we have gained the very ability to size down conventional radiowave antennas to the nanometer scale. Since nanoantennas provide an effective route to couple photons in and out of nanoscale volumes, they are considered as excellent tools to study and manipulate light-matter interaction at the nanoscale and have become an *indispensable* branch in a brand-new discipline, *plasmonics*, that concerns the physics of the interaction between electromagnetic fields and the collective oscillations of free electrons in a metal. A broad range of ground-breaking nanoantenna based

applications have been suggested spanning from the realm of life science, including biomedicine, bio-sensing, cancer treatment [1-4], to the research domain of spectroscopy [5-8], to a hot topic such as energy harvesting [9]. More global overviews of nanoantenna-based applications are given in many excellent review articles [10-11] and book (book chapters) [12-15].

Although similar to a classical antenna [16], the interaction of the current (the flow of the conduction electrons) in a nanoantenna with electromagnetic waves can be described by Maxwell's equations, there exists a fundamental difference. At microwave frequencies, the skin depth of the metals used is extremely small compared to the dimensions of the structure, several orders of magnitude. This means that they can be treated as structures carrying two dimensional surface currents, and the concept of surface impedance can be adopted. On the other hand, at optical frequencies the skin depth is comparable with the thickness of the nanostructures. Consequently, the current flowing in the structure is a three dimensional one. Any modeling scheme considered for these structures has to cope with this.

The traditional numerical techniques used in the microwave community are now being explored in the field of nanotechnology. Although most researchers use the differential approach, for example FDTD in [17-20], there are a few groups that have used the Method of Moments (MoM) to solve the integral equations describing the structure [21-22]. It can be proven that MoM can be implemented in a very efficient way for plasmonic nano-structures [23].

The modeling group around O. J. Martin [21-22] uses surface integral equations, where the boundaries of the

plasmonic structures are discretized. This is a classical technique, well-known in the microwave community. Another possibility is to use the volumetric equivalence principle. The nanoparticles are replaced by the equivalent induced electric polarization currents, and the volumetric integral equations describing the structure are subsequently solved by the Method of Moments (MoM). As a consequence of the fact that there, surface currents have to be described, this technique is rarely used in the microwave community. To our best knowledge, this approach was introduced to the plasmonics community in [23] and lies in the core of the present study.

This paper organizes as follows. In Section II, based on the classical electrodynamics, we theoretically formulate the interaction between light and a nanoantenna that is of an arbitrary shape and immersed in a generic environment composed of several dielectric regions. To numerically solve the problem, in Section III, the Volumetric Method of Moments is introduced [23-30]. In Section IV, to validate our implementation and demonstrate its capability of coping with nanoantennas of various geometries, we provide a vast amount of experimental data. Along the discussion, we also emphasize the link between the nanoantenna (array)'s linear and non-linear responses.

Although in principle any light – nanoantenna interaction can be solved by the formulation presented in Section III, the analysis is incident field dependent. In Section V, we focus on the eigenvalue problem of the present problem and extract the eigenvalues and eigenmodes of a nanoantenna. We demonstrate that the eigenmodes are not energetically orthogonal, to which the origin of Fano interference can be attributed. Moreover, we identify the material and geometry contributions to the modes' eigenvalues and demonstrate how they determine the nanoantenna's optical response.

Lastly, in Section VI, we push a step further: the (complex) natural frequencies and natural modes which are both incident and frequency independent are sought out for a nanoantenna. The link between the real and imaginary parts of the natural frequencies and the spectral position and quality factor a nanoantenna's surface plasmon resonances are thoroughly discussed. Further, the natural modes are demonstrated to be not energetically orthogonal, based on which the Fano resonance in a dolmen structure is explained.

II. THE INTERACTION BETWEEN A NANOANTENNA AND LIGHT

As shown in Fig. 1, at every space point, the total field is the sum of the scattered field and the incident field,

$$\mathbf{E}_{\text{tot}}(\mathbf{r}) = \mathbf{E}_{\text{scat}}(\mathbf{r}) + \mathbf{E}_{\text{inc}}(\mathbf{r}), \quad (1)$$

$\mathbf{r}$ is the observation point, that is, a point in the space. Especially at the position of the nanoantenna, there is a relation between the total electric field and the induced current (including both conduction and displacement currents),

$$\mathbf{J}_{\text{scat}}(\mathbf{r}, \omega) = j\omega(\varepsilon(\omega) - \varepsilon_0)\mathbf{E}_{\text{tot}}(\mathbf{r}, \omega). \quad (2)$$

Notice that ε_0 is the permittivity of the ambient background around the nanoantenna and $\varepsilon(\omega)$ is the dielectric function of metals. The isotropy and homogeneity of the material are assumed. The relation stated in Eq. (2) is the volume equivalence principle. Again, at the position of the nanoantenna, the scattered field is actually the electric field generated by the induced current,

$$\mathbf{E}_{\text{scat}}(\mathbf{r}, \omega) = -j\omega\mu_0 \int_{V'} \bar{\mathbf{G}}(\mathbf{r}, \mathbf{r}', \omega) \cdot \mathbf{J}_{\text{scat}}(\mathbf{r}', \omega) dv'. \quad (3)$$

By substituting Eq. (2) and Eq. (3) into Eq. (1), Eq. (1) becomes

$$\begin{aligned} & \frac{\mathbf{J}_{\text{scat}}(\mathbf{r}, \omega)}{j\omega(\varepsilon(\omega) - \varepsilon_0)} \\ &= -j\omega\mu_0 \int_{V'} \bar{\mathbf{G}}(\mathbf{r}, \mathbf{r}', \omega) \cdot \mathbf{J}_{\text{scat}}(\mathbf{r}', \omega) dv' + \mathbf{E}_{\text{inc}}(\mathbf{r}, \omega). \end{aligned} \quad (4)$$

Eq. (4) can be written more explicitly, with the unknown induced current $\mathbf{J}_{\text{scat}}$ on the left-hand side and the known incident field $\mathbf{E}_{\text{inc}}$ on the right-hand side,

$$\begin{aligned} & \frac{\mathbf{J}_{\text{scat}}(\mathbf{r}, \omega)}{j\omega(\varepsilon(\omega) - \varepsilon_0)} + j\omega\mu_0 \int_{V'} \bar{\mathbf{G}}(\mathbf{r}, \mathbf{r}', \omega) \cdot \mathbf{J}_{\text{scat}}(\mathbf{r}', \omega) dv' \\ &= \mathbf{E}_{\text{inc}}(\mathbf{r}, \omega). \end{aligned} \quad (5)$$

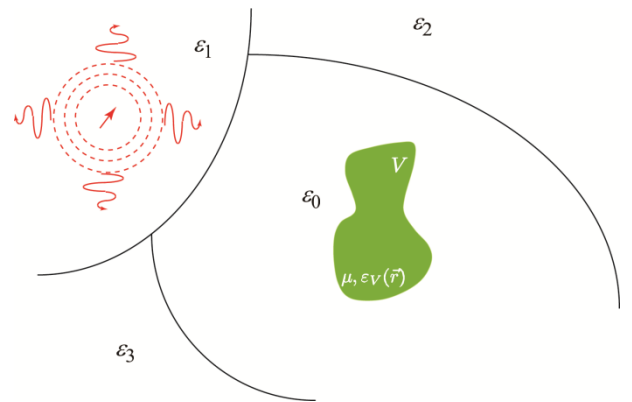


Fig. 1. An illustration of the interaction between light and a nanoantenna of arbitrary shape. A nanoscopic source which supports a current J radiates a wave that excites the nanoantenna described by a dielectric function $\varepsilon(\omega)$. The space is partitioned into several regions filled with various dielectrics with dielectric constant ε_i .

We cast Eq. (5) in an operator form,

$$Z|\mathbf{J}_{\text{scat}}(\mathbf{r}, \omega)\rangle = |\mathbf{E}_{\text{inc}}(\mathbf{r}, \omega)\rangle. \quad (6)$$

From the first term on the left hand side of Eq. (5), the total field operator is defined as,

$$\mathbf{E}_{\text{tot}}(\mathbf{r}, \omega) = Z_{\text{tot}}|\mathbf{J}_{\text{scat}}(\mathbf{r}, \omega)\rangle = \frac{\mathbf{J}_{\text{scat}}(\mathbf{r}, \omega)}{j\omega(\varepsilon(\omega) - \varepsilon_0)}, \quad (7)$$

The second term gives the scattered field operator,

$$\begin{aligned} -\mathbf{E}_{\text{scat}}(\mathbf{r}, \omega) &= Z_{\text{scat}}|\mathbf{J}_{\text{scat}}(\mathbf{r}, \omega)\rangle \\ &= j\omega\mu_0 \int_{V'} \bar{\mathbf{G}}(\mathbf{r}, \mathbf{r}', \omega) \cdot \mathbf{J}_{\text{scat}}(\mathbf{r}', \omega) dv'. \end{aligned} \quad (8)$$

By solving Eq. (5) or Eq. (6), the induced current $\mathbf{J}_{\text{scat}}(\mathbf{r}, \omega)$ is known and in principle the total electromagnetic fields at any spatial point can be obtained.

III. VOLUMETRIC METHOD OF MOMENTS (V-MoM)

Within the framework of a Volumetric Method of Moments algorithm (V-MoM) [23-30], we can discretize the continuous body of a nanoantenna (See Fig. 2(a)) by tetrahedral or hexahedral blocks (See Fig. 2(b)-(c)). The volumetric current flowing in two adjacent blocks is approximated by a three dimensional generalization of the well-known RWG rooftop functions [31,32], $\mathbf{f}_n(\mathbf{r})$.

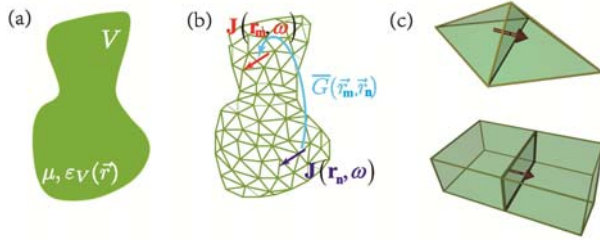


Fig. 2. The discretized representation of a nanoantenna. (a) The continuous body of an arbitrary nanoantenna; (b) An illustration of the discretization of the nanoantenna; (c) The tetrahedral and hexahedral blocks.

The induced current $\mathbf{J}_{\text{scat}}(\mathbf{r}, \omega)$ can be approximated by a weighted sum of these basis functions,

$$\mathbf{J}_{\text{scat}}(\mathbf{r}, \omega) = \sum_n j_n(\omega) \mathbf{f}_n(\mathbf{r}). \quad (9)$$

Inserting Eq. (9) into Eq. (5) gives one equation with n unknowns,

$$\sum_n j_n(\omega) Z|\mathbf{f}_n(\mathbf{r})\rangle = |\mathbf{E}_{\text{inc}}(\mathbf{r}, \omega)\rangle. \quad (10)$$

By further applying a testing procedure [33], that is, calculating the reaction of a “test function” $\mathbf{g}_m(\mathbf{r})$ to the electric field generated by a basis function $\mathbf{f}_n(\mathbf{r})$ and the incident field $\mathbf{E}_{\text{inc}}(\mathbf{r}, \omega)$,

$$\langle \mathbf{g}_m(\mathbf{r}), Z(\mathbf{f}_n(\mathbf{r})) \rangle = \int_{V_g} \mathbf{g}_m(\mathbf{r}) \cdot Z(\mathbf{f}_n(\mathbf{r})) dv', \quad (11)$$

$$\langle \mathbf{g}_m(\mathbf{r}), \mathbf{E}_{\text{inc}}(\mathbf{r}, \omega) \rangle = \int_{V_g} \mathbf{g}_m(\mathbf{r}) \cdot \mathbf{E}_{\text{inc}}(\mathbf{r}, \omega) dv'. \quad (12)$$

We approximate the infinitely dimensional operator in Eq. (6) by a finite N by N impedance matrix $\{z_{mn}\}$,

$$\{z_{mn}(\omega)\} \{j_n(\omega)\} = \{e_m(\omega)\}, \quad (13)$$

$$\begin{aligned} z_{mn}(\omega) &= \frac{1}{j\omega(\varepsilon(\omega) - \varepsilon_0)} \int_{V_g} \mathbf{g}_m(\mathbf{r}) \cdot \mathbf{f}_n(\mathbf{r}) dv \\ &+ j\omega\mu_0 \int_{V_g} \mathbf{g}_m(\mathbf{r}) \cdot \int_{V_f} \bar{\mathbf{G}}(\mathbf{r}, \mathbf{r}', \omega) \cdot \mathbf{f}_n(\mathbf{r}') dv' dv. \end{aligned} \quad (14)$$

In Eq. (13), $\{j_n(\omega)\}$ is a column vector which contains amplitudes of all basis functions and $\{e_m(\omega)\}$ denotes a column vector whose elements are the reaction of test functions to the incident electric fields as in Eq. (12)

Notice that in Eq. (12) and (14), the integrations are conducted with respect to the volumes of the *basis function* V_f and the *test function* V_g . By inverting the impedance matrix, the approximated induced current can be solved and the related physical quantities, e.g. far fields, can be evaluated. Nevertheless, before numerically solving Eq. (13), the elements in the impedance matrix have to be calculated, which involves two rather complicated steps, I) the evaluation of the Green's function; II) the evaluation of the double volumetric integral function. We refer the readers who are interested in the detailed implementations to several very well-written texts [24-26]. Please notice that in order to cope with the recently increasing interest in nanoscale antenna arrays (see many examples shown in the following section), the calculation of periodic Green's functions has been implemented on the basis of a quasi 3-D mixed potential integral equation formulation [34].

IV. VALIDATION OF THE V-MoM IMPLEMENTATION

In this section, a vast amount of experimental results is provided to validate our V-MoM implementation presented in

Section III. Also, by walking through these examples, we try to reveal a link between the linear and nonlinear optical response of a nanoantenna and suggest that Second Harmonic Generation (SHG) microscopy may be utilized as a powerful tool in imaging the near field of a nanoantenna (or nanoantenna array).

We start with a periodic rotary “G” shape structure [35,36] made of Nickel (Ni) with the Scanning Electron Microscope (SEM) image, depth profile, cell topology and dimensions shown in Fig. 3. By shining X polarized light onto the G structure, we can immediately receive a current pattern which exhibits four strong local electric field enhancements, i.e. “hotspots”, along the cell diagonal (See the red circled parts in Fig. 4). Note that in this section all the figures presenting the current distribution of a nanostructure are the plots of the root of the squared norm of the total current, that is, $\sqrt{\mathbf{J}^H \cdot \mathbf{J}} = \sqrt{|J_x|^2 + |J_y|^2 + |J_z|^2}$ where the operator H and $|x|$ represent the Hermitian transpose of a vector and the absolute value of a complex phasor, respectively. Compared with Noble metals (e.g. Gold, Silver, Copper), Ni suffers from a greater material loss at the wavelength of our interest: 800 nm. Hence, at the position of the most intense electric current concentrations, a larger joule heating effect (than the one generate by Noble metals) may be expected. The generated heat surpasses the melting point and consequently deforms the surface. This theoretical speculation can be immediately confirmed by the experimental observation in Fig. 4. There, atomic force microscopy (AFM) is made use of to demonstrate the hotspots “decorated” on the nanostructure.

Aside from the strongest hotspots, we can image other local current concentrations by further increasing the power of the

input pulse. Take the hotspots whose intensity is next to the strongest ones (See the dashed circled ones in Fig. 4(a)) as an example. Pumping the power of 1.23 mW, the AFM image unambiguously shows the imprints of these extra hotspots as accurately predicted by the simulations (See Fig. 4(c)). Further increases in the incident laser power imprint even more of the hotspots in Fig. 4(a). However, in this case the physical damage of the sample is unavoidable. Therefore, we can see that [35] this imprinting constitutes an experimental method for mapping near-field enhancements, which is easy to use, robust and offers a high resolution.

Further examples are the periodic Gold (Au) ninjastar structure [37] and the isolated Ni nanostripe structures [38] in Fig. 5 and Fig. 6. In Fig. 5(b), driven by the external field polarized along the horizontal direction at the wavelength of 800 nm, the positive and negative surface charges are separated at the tips of the star as highlighted by the dash red lines. Such a charge accumulation induces an intense current at the center position of the tips (See Fig. 5(d)), which generates strong enough heat to melt the sample. Bumps are formed as shown in the SEM and AFM images (See Fig. 5(f) and (h)). The same reasoning can be applied to the vertical polarization. Again, as demonstrated in Fig. 5(c), (e), (g), (i) and (k), the bumps denoted by the dashed lines in the SEM and AFM images match perfectly the positions of the “hotspots” in the simulated results. Notice that here the curved boundary of the Ninjastar structure is modeled by the hierarchical multilevel building block scheme proposed in [27] which is developed based on the volumetric nature of the current flowing in a nanoantenna.

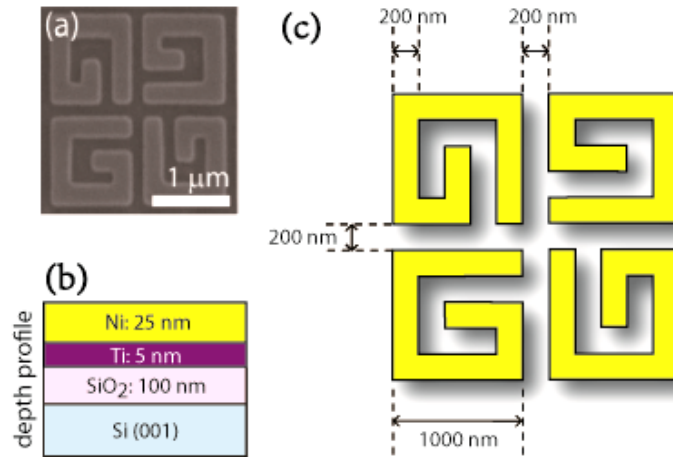


Fig. 3. The Scanning Electron Microscope (SEM) image (a), depth profile (b), cell topology and dimensions (c), periodic rotary “G” shape structure. The period is 2400 nm.

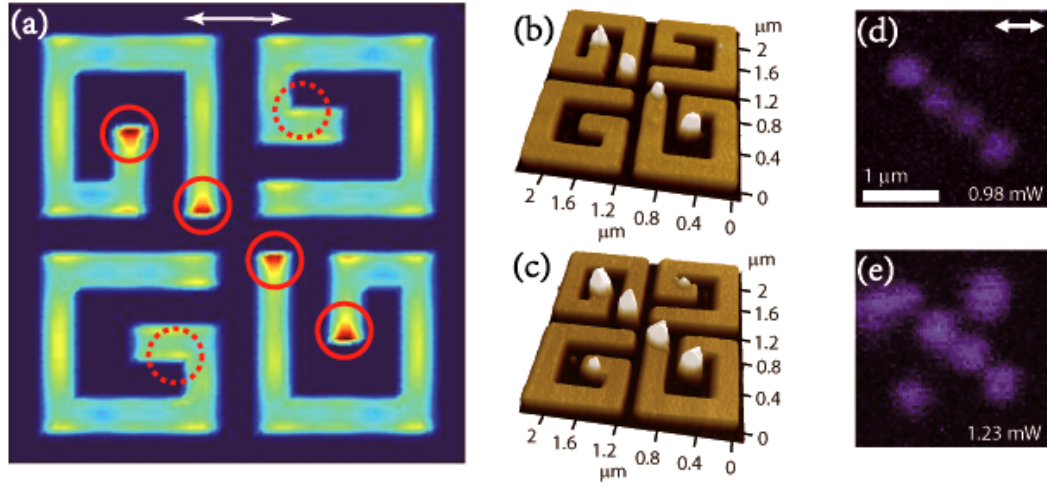


Fig. 4. Illustration of the simulated electric current, the “decorated” bumps and the SHG hotspots. In (a), the electric currents at the fundamental frequency are collected at 12.5 nm under the surface and encoded from the blue to the red color to denote the current intensity from the weakest to the strongest. The polarization of the incident wave is specified by the white arrow. The most intense current localization are marked by the red full circles, while the hotspots next the most intense ones are highlighted by the red dashed circles. In the simulation, the incident wave is at 800 nm and all the layer structure except the 5 nm Titanium (Ti) layer in Fig. 3(b) is taken into account. In (b), after the illumination of laser beam with 0.98 mW at 800 nm, the samples are scrutinized by the atomic force microscopy (AFM). In (c), four bumps are well aligned with the theoretical prediction at the fundamental frequency, i.e. the “hot” spots in the solid red circles. After increasing the power to 1.23 mW, extra bumps are imprinted on the sample surface which match the next intense points (red dashed circles) in (a). In (d) and (e), the corresponding Second Harmonic (SH) signals are collected by the Second Harmonic Generation (SHG) microscopy. A clear link between the fundamental electric current, laser imprints and SHG emission can be established.

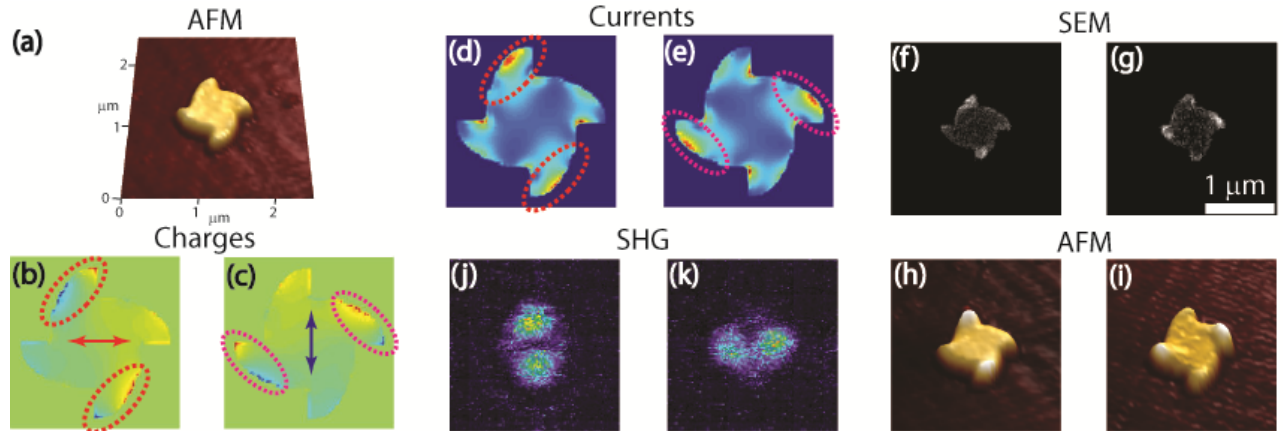


Fig. 5. Illustration of the simulated surface charge and electric current, the SEM and AFM images of the “decorated” samples and the SHG near field images. The AFM image of the ninjastar structure before experiments is shown in (a). The depth profile is the same as the one present in Fig. 3(b) except the Ti layer is replaced by a Chromium (Cr) layer. The period of the structure is 2400 nm. In (b) – (e), the surface charges and electric currents taken at 12.5 nm under the surface are shown. The electric charge is coded from the blue color to the red color to represent the negative and the positive charges, while the electric current is presented in the same way as in Fig. 4(a). The polarizations of the incident light used in the simulations are specified by the horizontally and vertically placed arrows. The simulations are conducted at 800 nm. All the layers except the Cr layer are taken into account in the simulations. The “hotspots” in the simulations are denoted by the dashed red and magenta circles for different polarizations. In (f) and (g) the SEM image, in (h) and (i) the AFM image, and in (j) and (k) the SHG emission, the “decorated” bumps and their corresponding SHG hotspots match the simulated results in an excellent way.

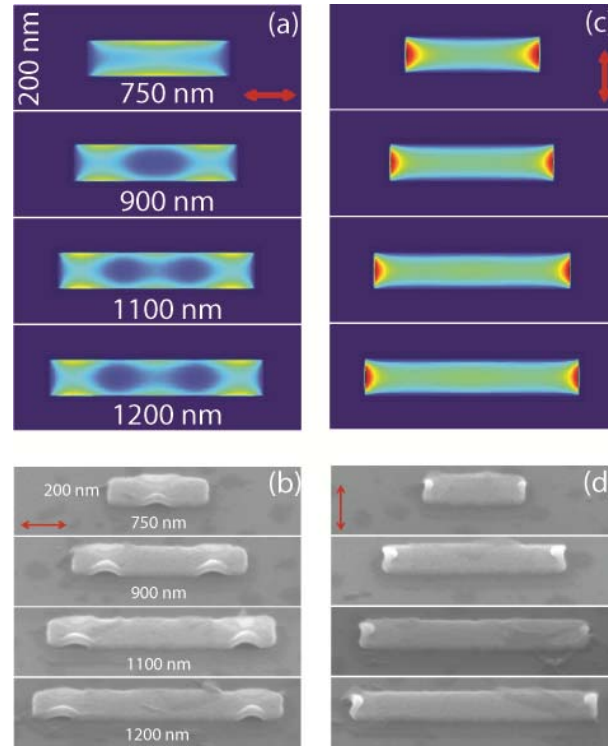


Fig. 6. Illustration of the simulated electric current and the SEM images of the “burnt” bar-shape samples. The nanobars are with various lengths (750 nm – 900 nm – 1100 nm – 1200 nm), fixed width (200 nm) and a thickness of 35 nm. The depth profile of the structure is the same as the rotary G structure shown in Fig. 3(b) except the Ti layer is replaced by a Cr layer. In the simulations (a) and (c), all the layer structure is taken into account. The polarizations are shown by the red arrows. In (b) and (d), for clarity, the SEM images are taken at the angle of 45 degree. Again, a good agreement can be observed between the simulated and experimentally obtained results.

For the nanostripe [38] structures, we fix the incident wavelength at 800 nm and vary the stripe’s lengths. The simulated currents in Fig. 6(a) exhibit that under the excitation of a horizontally polarized field, the longitudinal electric current modes with different number of antinodes (maxima) are excited. In contrast, when the incident field is polarized in the vertical direction, since the width is too short compared with the wavelength, only the lowest order transversal mode is excited. Again, as expected, the SEM images performed at 45 degrees unambiguously demonstrate the excellent match between the local current maxima and the pattern of bumps.

More importantly, the physical mechanism behind the formation of the bumps can be intuitively understood by a hydrodynamic process [37]: when a water droplet impinges on a surface of water, a column of water shoots upwards and falls back in (See the left panel in Fig. 7(a)). A similar process also applies to nanoantennas (See the schematic description in the right panel of Fig. 7(b)). We illuminate a G-shape sample with a single light pulse of 60 femtosecond (10^{-15} s) duration. When the sample is shined by a horizontally polarized laser pulse with a power of 30 mW, as can be seen from the SEM image in Fig. 7(f) – (k), two bumps appear at the ends of the G shape spiral line. As the laser power increases, the bumps intend to detach from the sample (See the red dashed circle in Fig. 7(i)), form a “water” column with a metal “droplet” on top of it (See the red dashed circle in Fig. 7(j)) and finally land back on the sample (See the red dashed circle in Fig. 7(k)). Nevertheless, it is worth noticing that here we must recognize that the “water droplet” explanation is maybe too simple. One

can argue that the power delivered by the femtosecond laser and thus the induced temperature in a single Nickel layer is not high enough to melt the metals [39-45]. The melting might be due to the oxidation effects (which can be thermally violent) of the Ni – Ti layer (in Fig. 3(b)) upon the arrival of the femtosecond laser pulse. Such a speculation clearly points out the direction of possible future work: i) experimental work that excludes all possible chemical factors altogether; and ii) a further development of our solver to include thermodynamical effects.

Since the local electric current enhancements are large enough to trigger (possibly, an oxidation induced) melting of the metals and the high surface to volume ratio of a planar nanostructure promises the breaking of the inversion symmetry, it is then natural to think of the second harmonic response originating from these hotspots. As shown by the simulated charge distributions in Fig 7(b), the phenomenon of electric charges driven to the edges by the electric field resembles a harmonic oscillator. When the incident light is not very strong, the charges linearly move back and forth in accordance with the incident light and constitute an electric current. Especially, within the hotspots (See the reddest points in Fig. 7(c)), the electric currents become most intense. As the intensity of the incident field increases, the oscillators at the “hotspots” start to enter the nonlinear regime. The corresponding electric currents radiate at the second harmonic (SH) and SHG microscopy is able to detect the generated SH signal (See the “hotspots” in Fig. 7(d)). Similar mechanisms can be observed for the previously analyzed nanostructures.

In Fig. 4(d,e) – Fig. 5(j,k), excellent matches between the simulated results, the SEM/AFM images and the SHG images are observed. Therefore, a strong link can be established between the local electric current enhancements at the fundamental frequency and the “hotspots” in the SHG images.

In all the aforementioned examples, linearly polarized light that separates charges in a specific direction is always employed in the illumination of the samples. In contrast, for circularly polarized light, the electric field changes its direction in a rotary manner and consequently is capable of driving the charges around a nanostructure. Such a concept provides an elegant way to control the surface charge flow and may allow the design of plasmonic circuit elements. Take a closed loop, i.e. a square-ring structure [46] for example (See the topology, dimensions, and depth profile in Fig. 8(a) and (b)), is considered. For linearly polarized illumination, two strong hotspots have been observed in the SHG image and, as anticipated, correspondingly two strong local electric accumulations in the numerical calculations (See Fig. 8(c) and

(e)). On the contrary, for circularly polarized light, the surface charge is spread over the entire structure, see the more homogeneous pattern obtained in the numerical and experimental results in Fig. 8(d) and (f). Then, for a U structure [47] two paths are designed for the surface charge flows. As the left (right) circularly polarized light is applied, the surface charge is rotated to follow the upper (bottom) path, reach and finally accumulate at the end of the path. This concept can be unambiguously confirmed by the SHG images in Fig. 9(d,e) from which it can be seen that the hotspots at point A and B (white filled parts in Fig. 9(d,e)) can be selectively excited by the circularly polarized light. Further, as expected, the vertically polarized light is well-coupled with the U-shape structure so that in this case both A and B points are bright in contrast to the two dark branches when the horizontally polarized light is applied. Based on the above observations, a U-shape four-phase “switch” can be conceived and may become an indispensable element in future plasmonic circuits.

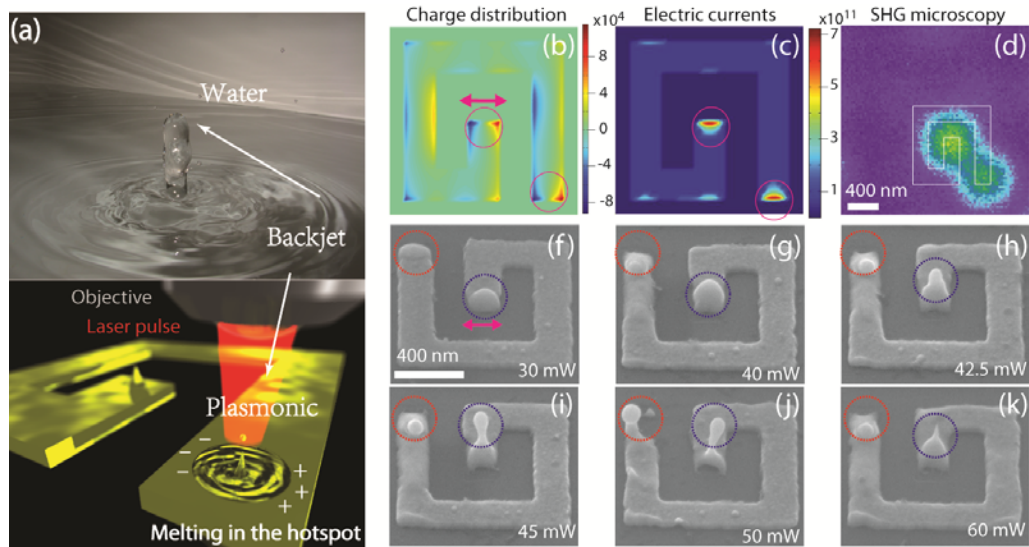


Fig. 7. Illustration of the physical mechanism behind the formation of the bumps. In response to the horizontally polarized light, the surface charges, e.g. the red circles in (b), are separated to the structure’s edges and form an oscillator. The corresponding current distribution is shown in (c). With the increasing laser intensity, the charge oscillations enter the nonlinear regime and consequently the SH emission can be detected in (d). Moreover, at the local current enhancements, due to the ohmic loss, the local temperature surpasses the metal’s melting point and consequently allows the occurrence of the hydrodynamic process as illustrated in (a). Such a process can be clearly traced through the observation of (f) – (k). Take the outer end of the G-shape spiral, i.e. the part highlighted by the red dashed circles, as an example. With increasing laser power, a bump forms, grows, detaches and lands the samples.

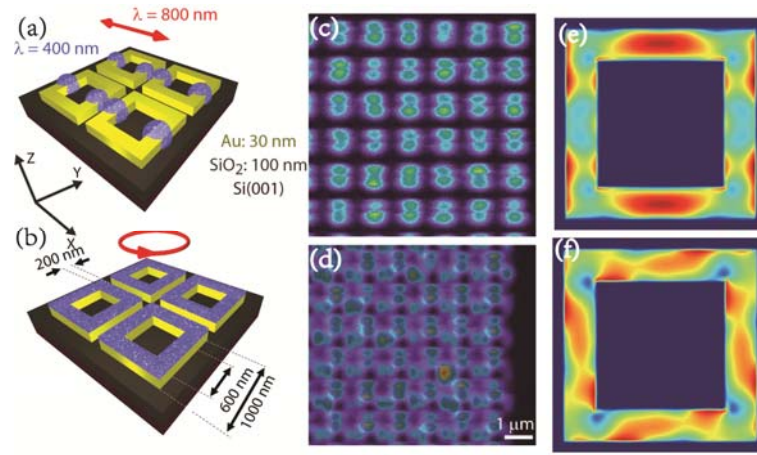


Fig. 8. Illustration of homogeneously distributed surface charges and optical fields. (a) and (b) show the topology, dimensions and the depth profile of the periodic square ring structure. The period is 1200 nm. We illuminate the sample from the top with an incident light at 800 nm. Since the square ring is a highly symmetric structure, here we only investigate the effects of Y polarized and left polarized excitations. Upon the Y-polarized excitation, in (c,e) the most intense local electric currents enhancements and accordingly the strongest SH signals are observed along the center line of the square ring. For the left-polarized excitation, in (d,f) the current pattern and the SH emission exhibit a more homogeneously distributed pattern.

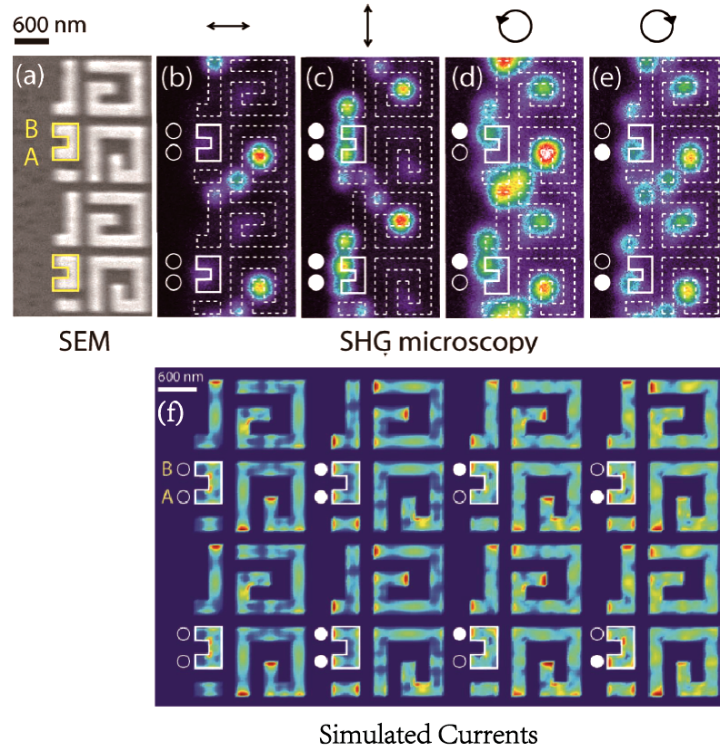


Fig. 9. Illustration of a U-shape four-phase switch. The SEM image of the U shape structure is shown in (a). The “contacts” of the switch are denoted by “A” and “B”. In both experiments and simulations, the presence or absence of hotspots at “A” and/or “B” points is indicated by full or empty white circles. We illuminate the sample from the top with an incident light at 800 nm with different polarizations. On one hand, for the X polarized incident light, both “contacts” are dim as shown in (b) and the first column in (f), which is practically equivalent to a “off” state, while as illustrate in (c) and the second column in (f), for the Y polarized light, points “A” and “B” are bright, which thus forms a full “on” state. On the other hand, as in (d,e) and the third and fourth columns in (f), the left (right) circularly polarized can excite the upper (bottom) path. In this case, the switched is selectively “wired” to the “A” point or the “B” point. Therefore, a polarization sensitive four-phase switch is realized. (b) – (e) demonstrate the collected SH signals. The colors in these panels are coded from purple through blue, green, yellow, red and white to designate the intensities. For clarity, the contour of the underlying structure is superposed onto the SHG images and delineated by white dash lines. The corresponding simulated currents are demonstrated in (f).

V. EIGENMODE ANALYSIS OF NANOANTENNAS

As discussed in Section II, Eq. (6) includes all the physics in the light-nanoantenna interaction. In principle, based on Eq. (6) and provided the incident electric field is given (e.g. a plane wave or the fields emitted by a fluorescent molecule), the induced polarization currents and the associated scattered fields can be solved, which seems to put an end to the story. Nevertheless, the full-wave solution to Eq. (6) is *incident field dependent*, which might in some cases (e.g. the complex current distributions of the G structure) blur the physical picture in both the analysis and design of a nanoantenna. In this section, we will pursue an incident field independent solution, i.e. the eigenmodes of a nanoantenna, and reveal their distinct roles in determining antenna's response. We assume the eigenvalue problem for Eq. (6) exists and is defined as [48],

$$Z|\mathbf{J}_n(\mathbf{r}, \omega)\rangle = \lambda_n(\omega)|\mathbf{J}_n(\mathbf{r}, \omega)\rangle. \quad (15)$$

Note that as in [48], here we define the eigenmodes of a nanoantenna based on the impedance operator Z . However, we have to point out that this definition is quite different from the usage of the terminology “eigenmode” (or “normal mode”) in the physics community, especially the quantum optics community. There, the word “eigenmode” (or “normal mode”) is especially reserved for the free space modes which are real and orthogonal in an inner product sense and based on which the electromagnetic field is quantized [49].

Since the total field operator Z_{tot} is a scalar, we can distinguish two different contributions to the eigenvalue (similar to the case of a dielectric antenna [50]),

$$\lambda_n(\omega) = \lambda_{n,mat}(\omega) + \lambda_{n,scat}(\omega), \quad (16)$$

where $\lambda_{n,mat}(\omega) = \frac{1}{j\omega(\varepsilon(\omega) - \varepsilon_0)}$ is the material contribution and $\lambda_{n,scat}(\omega)$ is controlled by the geometry of the nanoantenna. Then the total field operator can be eliminated on both sides of Eq. (15),

$$Z_{scat}|\mathbf{J}_n(\mathbf{r}, \omega)\rangle = \lambda_{n,scat}(\omega)|\mathbf{J}_n(\mathbf{r}, \omega)\rangle, \quad (17)$$

From Eq. (17), it can be immediately seen that the eigenmodes $|\mathbf{J}_n(\mathbf{r}, \omega)\rangle$ are actually *independent from the material of the nanoantenna*.

Due to the reciprocity of the Green's function, Z_{scat} is a *complex symmetric* operator and the eigenmodes are complex function and orthogonal in a pseudo inner product sense [51],

$$\langle \mathbf{J}_m(\mathbf{r}, \omega), \mathbf{J}_n(\mathbf{r}, \omega) \rangle = \begin{cases} 1 & m = n \\ 0 & m \neq n \end{cases}. \quad (18)$$

Further, we can define the right hand side of Eq. (15) as an “eigen” incident field $|\mathbf{E}_{inc,n}(\mathbf{r}, \omega)\rangle$ for the n^{th} eigenmode,

$$|\mathbf{E}_{inc,n}(\mathbf{r}, \omega)\rangle = \lambda_n(\omega)|\mathbf{J}_n(\mathbf{r}, \omega)\rangle. \quad (19)$$

By evaluating the work done by the n^{th} “eigen” incident field $|\mathbf{E}_{inc,n}(\mathbf{r}, \omega)\rangle$ on the m^{th} eigenmode $|\mathbf{J}_m(\mathbf{r}, \omega)\rangle$, the energetic coupling between eigenmodes can be found. The *self-impedance* of the n^{th} eigenmode $z_{nn}(\omega)$ and the *mutual impedance* between the m^{th} eigenmode and the n^{th} eigenmode $z_{mn}(\omega)$, can be defined as,

$$\begin{aligned} z_{mn}(\omega) &= \langle \mathbf{J}_m^*(\mathbf{r}, \omega), \mathbf{E}_{inc,n}(\mathbf{r}, \omega) \rangle \\ &= \langle \mathbf{J}_m^*(\mathbf{r}, \omega), \lambda_n(\omega) \mathbf{J}_n(\mathbf{r}, \omega) \rangle \\ &= d_{mn}(\omega) \lambda_n(\omega), \end{aligned} \quad (20)$$

Note that in Eq. (20), the factor $1/2$ is systematically dropped and d_{mn} is a unitless factor taking into account the fact that the eigenmodes are not orthogonal in the inner product sense, i.e., not *energetically orthogonal*.

When an incident field is applied, the induced current $\mathbf{J}_{scat}(\mathbf{r}, \omega)$ can be written as a linear combination of the eigenmodes,

$$\mathbf{J}_{scat}(\mathbf{r}, \omega) = \sum_n i_n(\omega) \mathbf{J}_n(\mathbf{r}, \omega), \quad (21)$$

where $i_n(\omega)$ is the current amplitude of the n^{th} eigenmode,

$$i_n(\omega) = \frac{\langle \mathbf{J}_n(\mathbf{r}, \omega), \mathbf{E}_{inc}(\mathbf{r}, \omega) \rangle}{\lambda_n}. \quad (22)$$

Further, the voltage $v_n(\omega)$ felt by the n^{th} eigenmode can be defined as,

$$v_n(\omega) = \langle \mathbf{J}_n^*(\mathbf{r}, \omega), \mathbf{E}_{inc}(\mathbf{r}, \omega) \rangle. \quad (23)$$

By collecting Eq. (20), (22) and (23), we can reconstruct the operator Z in Eq. (6)

$$\{z_{mn}\}\{i_n\} = \{v_m\}. \quad (24)$$

Notice that Eq. (24) is akin to the Volumetric method of Moments (V-MoM) formulation in Eq. (13). Instead of using subdomain basisfunction as in Eq. (13), Eq. (24) employs the antenna's eigenmodes as entire domain basis functions and reformulates the Z operator in such a way that its diagonal elements, i.e. self impedance of an eigenmode, describe the self energetic coupling of eigenmodes and are a factor related to the eigenvalue; its off diagonal elements, i.e. mutual impedance between eigenmodes, measure the power transferred between ports (eigenmodes) and emphasize the

fact that the eigenmodes are not necessary to be energetically orthogonal, i.e. the modes are coupled. When an incident field is applied, the amplitude and the phase of the coupling coefficient and the input power are determined for each eigenmode as in Eq. (22) and Eq. (23). Alternatively, Eq. (24) can be interpreted as an N-port network (See Fig. 10). Each port represents a *material independent* eigenmode and interacts with the “outside” world. Ports can be excited or “open-circuited” depending on how the eigenmode “interfaces” with the excitation. Since the eigenmode is equated with the port, for the rest of this section, we use these two words interchangeably.

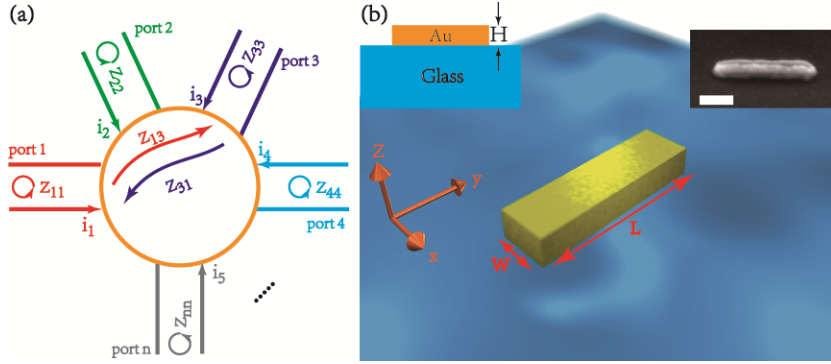


Fig 10. (a) Schematics of the equivalent N port network. The topology, the depth profile, and the SEM image of the nanobar ($L = 370$ nm, $W = 70$ nm, $H = 50$ nm) are presented in (b). Throughout the article, the white bar represents 100 nm.

Lastly, the self-coupling power of the n^{th} eigenmode and the mutual coupling power between the n^{th} and m^{th} eigenmodes can be evaluated as,

$$p_{nn} = z_{nn} |i_n|^2, \quad (25)$$

$$p_{mn} = i_m^* z_{mn} i_n. \quad (26)$$

The power p_m delivered to the m^{th} eigenmode is,

$$p_m = \sum_n i_m^* z_{mn} i_n. \quad (27)$$

The N-port network interpretation of a nanoantenna can be unambiguously confirmed by examining the nanobar structure. The first three eigenmodes, their corresponding self-impedances and the coupling coefficients under Y polarized light incident along Z direction (see Fig. 11) are evaluated for a gold nanobar of 370 nm by the V-MoM formulated in Section II. The extinction cross sections are calculated by the V-MoM and Finite Difference Time Domain (FDTD) methods [52]. The description of the topology and the dimensions of the structure can be found in the caption of Fig. 10. As shown in Fig. 11(c), because of the symmetry of the L2 mode with two charge nodes, it is decoupled from the normally incident light and consequently the current amplitude of the L2 mode is zero. That is, the L2 port is effectively “open-circuited”. For the excited L1 and L3

modes, as the imaginary part of the self-impedances crosses zero, the current amplitudes reach the maximum, with a small but noticeable spectral difference, as indicated by the grey dashed lines through Fig. 11(b)-(e). This spectral difference arises from the real part of the self-impedance. Accordingly, the real part of the power at the port, i.e. the power lost via the dissipation and the radiation, is maximized. Thus, a corresponding resonance is observed in the extinction cross section spectra, as demonstrated numerically by Fig. 11(d) and experimentally by Fig. 11(e).

However, the role of the mutual impedance, which describes the interaction between ports, should not be overlooked. It can be seen in Fig. 11(f)-(g) and Eq. (20) that the N-port network equivalence proposed in this work is not a “reciprocal” network defined in the classical microwave network theory, where $z_{mn} = z_{nm}$ [53]. The discrepancy arises due to the complex symmetric nature of the field operator in Eq. (15). The eigenmodes belonging to a complex symmetric operator, if they exist, are complex functions and only complex orthogonal as in Eq. (18), i.e. Eq. (18) only assures the reciprocity $\int_{V'} \mathbf{J}_1 \cdot \mathbf{E}_2 dv' = \int_{V'} \mathbf{J}_2 \cdot \mathbf{E}_1 dv' = 0$ defined based on the concept of reaction [54] but doesn't guarantee any reciprocity of the related power $\int_{V'} \mathbf{J}^* \cdot \mathbf{E} dv'$ and accordingly

the impedance. For a conventional microwave network, the reciprocity of an N-port network is based on the realness of

the transverse modal fields at each port. If the realness is assumed, the concept of reaction and the concept of power coincide. Consequently, z_{mn} is equal to z_{nm} , suggesting a conventional “reciprocal” network. Moreover, the mutual-impedance between ports shapes the power at each port. To show this we take the L3 mode as an example. Since the L2 mode is not excited, there is no power transfer between the L2 mode and the L3 mode. Yet, the L1 mode is excited and coupled with the L3 mode. Especially around the resonance of the L3 mode, a negative power flow from the L1 to L3 mode is observed (Fig 11(i)), suggesting that part of the work done by the incident field on the L3 mode, instead of

being radiated by the L3 mode, is withdrawn by the L1 mode. By adding both the self-coupling and mutual coupling contribution, the power at the L3 port shows an asymmetric line shape, as indicated in Fig 11(j)-(k), marking the occurrence of a Fano interference [55]. It is also worth noticing that for spheres the complex symmetric operator in Eq. (15) reduces to a normal operator whose eigenmode and their corresponding fields, i.e. the multipoles employed in a Mie expansion, are energetically orthogonal. Hence, the mutual coupling between eigenmodes disappears and Fano-like interference is absent in spherical nanoparticles.

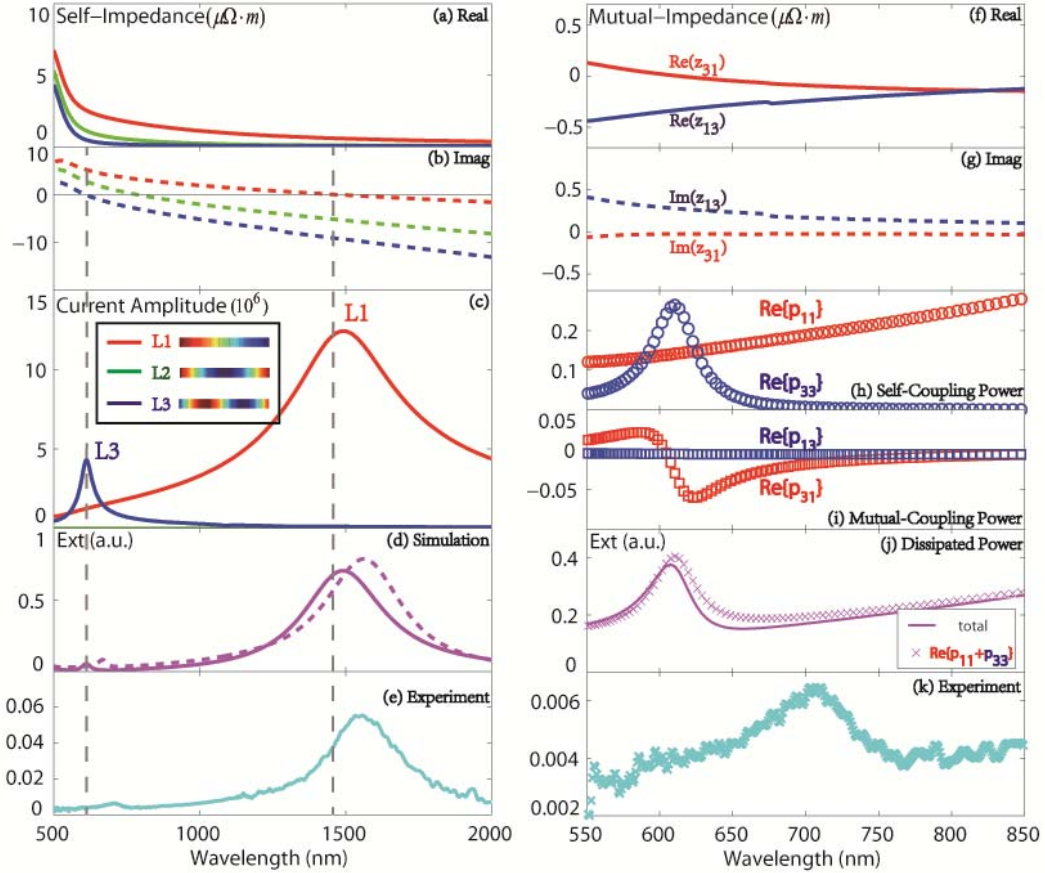


Fig. 11. In panels (a) and (b), the real (solid) and imaginary parts (dashed) of the self-impedances for L1 (red), L2 (green) and L3 (blue) eigenmodes (as shown in the inset of (c)) are presented. The coupling coefficients for L1, L2 and L3 under Y-polarization illumination are illustrated in (c). (d) and (e) are the simulated (V-MoM – solid magenta line and FDTD – dashed magenta line) and measured extinction cross sections (cyan). In panel (f) and (g), the real (solid) and imaginary parts (dashed) of the mutual-impedance between port 1 and port 3 are shown. The real part of mutual coupling power (squares) together with the self-coupling power (circles) is shown in (h) and (i). In (j), a comparison is made between a direct sum of self-coupling power (crosses), i.e. p_{11} and p_{33} , and the total power (solid). The asymmetric lineshape can be also seen in the measured extinction cross-section (k). In the inset of (b), the colors are coded from blue to red to denote the negative and positive charge accumulations.

VI. NATURAL MODE ANALYSIS

It is worth noticing that the eigenmodes extracted in Section V are *incident field independent*, they are still frequency dependent. To see this, a V-shape nanoantenna [56] is taken as an example (See the topology, the depth profile and the SEM

image in Fig. 12). In the framework of V-MoM algorithm, we extract the eigenmodes and evaluate their corresponding far field distributions at 500 nm and 2500 nm (See the surface charge distribution and their far field patterns in Fig. 13). Notice that in the simulation of the far fields the substrate is not taken into account. It can be readily seen from Fig.

13(c)-(d) that at 500 nm, the far field pattern shows a “donut” shape; however, at 2500 nm, the pattern is completely deformed. The substantial change in far fields can be traced back to the corresponding mode distribution (See the circled parts in Fig. 13(a)-(b)).

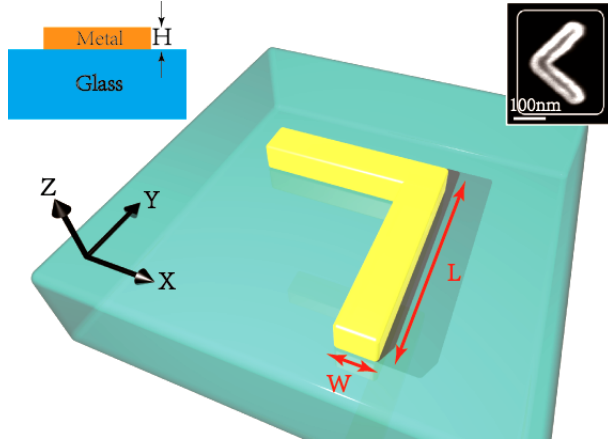


Fig. 12. Illustration of an Au V-shape nanoantenna. The dimensions of the V structure are $L = 250$ nm, $W = 70$ nm, $H = 50$ nm and the angle between two arms is 90 degrees. The depth profile, the SEM image and the coordinate system are shown in the insets. The white bar represents 100 nm.

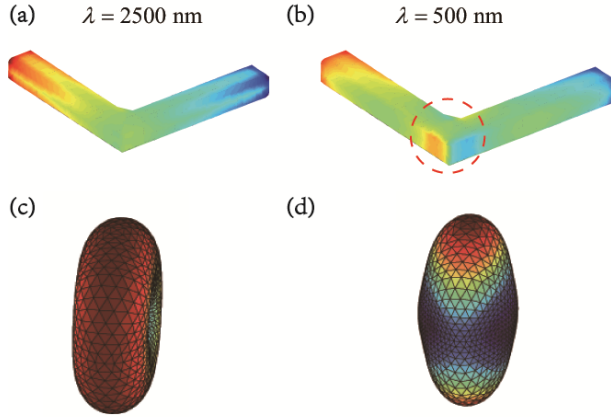


Fig. 13. Illustration of eigenmodes' frequency dependency. Two wavelengths are considered: the first column 2500 nm and the second column 500 nm. In (a) and (b), the surface charge distributions are plotted. The charge is coded from blue to red to denote the negative and positive charge accumulations. In (c) and (d), the corresponding far field intensity $|E|^2$ (without considering the substrate effects) is illustrated with the blue color representing the smallest intensity and the red color the largest.

Indeed, depending on the geometry, material and boundary condition, a mechanical system, e.g. a bridge, drum, waveguide, molecule, or an atom, has a set of natural (normal) modes. Such a set together with the corresponding natural frequencies characterizes the most intrinsic aspects of the system and determines the system's free and forced response. In electromagnetics, the interests in natural frequencies and natural current modes of a general radiating structure, e.g. microwave scatterers or antennas, arose from the studies on the interaction of a nuclear electromagnetic pulse (EMP) with metallic objects in early 1970s [57]. There, scatterers' transient response universally exhibits damped sinusoids, which suggests the existence of natural frequencies in the complex frequency domain where the real and imaginary axes represent the oscillation frequency and the decay rate respectively. In this section, we introduce a rigorous full wave natural mode analysis to the optical spectrum and push one step forward to find a mode that is *both frequency and excitation independent* by solving the following equation [57],

$$Z[\mathbf{J}_\alpha(\mathbf{r}, \omega_\alpha)] = 0. \quad (28)$$

In Eq. (28), $\mathbf{J}_\alpha(\mathbf{r}, \omega_\alpha)$ is the α^{th} natural mode current whose natural frequency is ω_α . By utilizing the V-MoM algorithm, we can numerically solve Eq. (28) by seeking for the solution of Eq. (13) when the excitation is zero,

$$\{z_{mn}(\omega_\alpha)\} \{j_n(\omega_\alpha)\} = \mathbf{0}. \quad (29)$$

That is, the determinant of the impedance matrix is zero,

$$\det(\{z_{mn}(\omega_\alpha)\}) = 0. \quad (6.30)$$

According to the above-discussed procedure, the natural frequencies and natural modes of gold nanorods with different lengths are numerically calculated with the V-MoM code. The dimensions, materials, mesh sizes and surroundings of the nanorods are tabulated in Table 1. The calculated natural frequencies are shown in Fig. 14(a) and tabulated in Table 2. The calculated natural modes of the 300 nm nanorod are shown in the inset of Fig. 14(a). For clarity, the factor 2π is dropped from all the real parts of the natural frequencies.

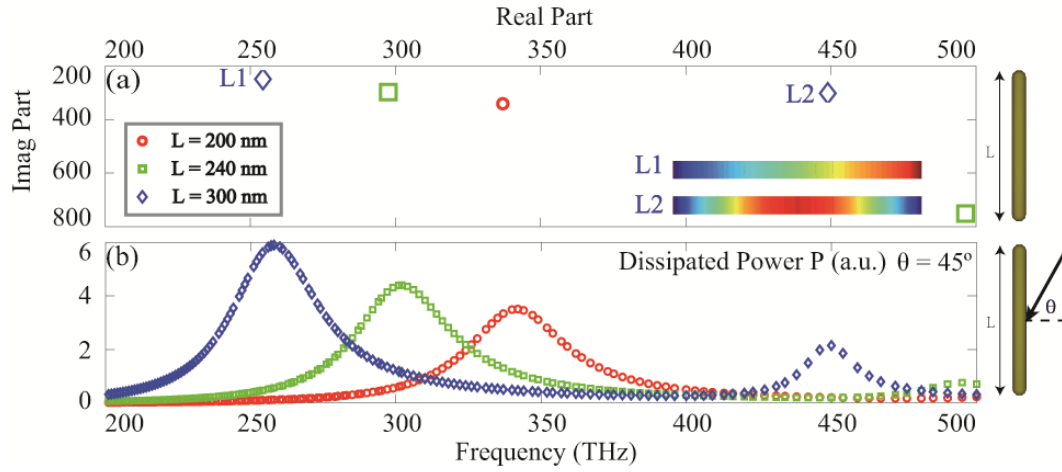


Fig. 14. Natural frequencies of nanorods determine the line positions of surface plasmon resonances. (a) presents the complex natural frequencies of nanorods of different lengths. The real and imaginary part of the natural frequencies has the unit of THz and Tera-radians per second respectively. The top surface charge distributions of the first two normal modes (L1 and L2) are shown in the inset for the case $l = 300$ nm. The surface charge is coded from blue to red to denote the negative and positive charge accumulations. In (b), the dissipated power is evaluated for an incoming wave with incidence angle 45° with its polarization perpendicular to the direction of propagation and lying in the incidence plane.

Parameters	Nanorods		
Length (L)	200 nm	240 nm	300 nm
Width (W)	40 nm	40 nm	40 nm
Thickness (T)		40 nm	
Material		Gold	
Surrounding		Vacuum	
Mesh cell sizes	20 nm x 20 nm x 20 nm		

Table 1. Parameters used in the simulations. Note that the materials properties of Gold in the complex frequency plane is approximated the corresponding real frequency axis behavior, i.e. $\varepsilon(\text{Re}(\omega))$.

	L1	L2
200 nm	336.990 + 341.224i	556.564 + 2830.428i
240 nm	297.565 + 297.780i	497.472 + 751.535i
300 nm	254.355 + 249.281	448.910 + 302.259i

Table 2. Natural frequencies of 200 nm, 240 nm and 300 nm nanorods. The real part is with the unit of THz, while the imaginary part is with Tera-radians per second.

When an external excitation of frequency ω is applied, the polarization current is solved from Eq. (13)

$$\begin{aligned} \{j_n(\omega)\} &= \{\mathbf{z}_{mn}(\omega)\}^{-1} \cdot \{e_m(\omega)\} \\ &= \frac{\text{adj}(\mathbf{z}_{mn}(\omega))}{\det(\mathbf{z}_{mn}(\omega))} \cdot \{e_m(\omega)\}. \end{aligned} \quad (31)$$

$\text{adj}(\mathbf{z}_{mn}(\omega))$ is the adjugate matrix of the impedance matrix.

The determinant of the impedance matrix $\det(\mathbf{z}_{mn}(\omega))$ is a polynomial that can be decomposed into factors $(j\omega - j\omega_\alpha)^n$. $\omega_\alpha = \omega_{\alpha r} + j\omega_{\alpha i}$ is a natural frequency and n is the order of the natural frequency, indicating the degree of degeneracy of the natural modes. We assume for a nanorod only the first order natural frequencies ($n = 1$) are present and the response can be re-written as [57]

$$\begin{aligned} \{j_n(\omega)\} &= \sum_{\omega_\alpha} \frac{\mathbf{R}_\alpha}{(j\omega - j\omega_\alpha)} \cdot \{e_m(\omega)\} \\ &= \sum_{\omega_\alpha} \frac{\{j_n(\omega_\alpha)\} \{j_n(\omega_\alpha)\}^T}{(j\omega - j\omega_\alpha)} \cdot \{e_m(\omega)\} \\ &= \sum_{\omega_\alpha} \frac{\{j_n(\omega_\alpha)\}^T \cdot \{e_m(\omega)\}}{(j\omega - j\omega_\alpha)} \cdot \{j_n(\omega_\alpha)\}. \end{aligned} \quad (32)$$

$\mathbf{R}_m$ is the system residue matrix at each natural frequency. It has been shown that $\mathbf{R}_m$ is a dyadic and can be decomposed as the outer product of natural mode $\mathbf{j}_m$ and its transpose $\mathbf{j}_m^T$. For light of frequency ω around $\omega_{\alpha r}$, by neglecting the contribution from the α^{th} natural frequency with a negative

real part, we can *approximate* the amplitude of $\{j_n(\omega)\}$ (rigorously, such a approximation can be only made when the loss is small),

$$|\{j_n(\omega)\}| \propto \frac{1}{\sqrt{(\omega - \omega_{ar})^2 + \omega_{ai}^2}}. \quad (33)$$

It is readily seen from Eq. (33) that when ω is around ω_{ar} , the response reaches a maximum. Therefore, the positions of the resonances can be correctly predicted by the real parts of the natural frequencies, as shown by the line positions of the resonances in Fig. 14 (a) and (b).

Moreover, a modal quality factor Q_{mod} can be defined for each natural mode. Consider an incoming electromagnetic pulse $\delta(t)$, which contains all frequency components with the same amplitude 1. Eq. (32) leads to

$$\{j_n(t)\} = \sum_{\omega_\alpha} K_{\omega_\alpha} \{j_n(\omega_\alpha)\} e^{j\omega_\alpha t}. \quad (34)$$

K_m is defined as the coupling coefficient of the α^{th} natural mode, which is a constant independent of frequency. In Eq. (34), all natural modes are present. By noticing that

$$j\omega_\alpha t = -\omega_{ai}t + j\omega_{ar}t, \quad (35)$$

natural mode oscillates with frequency ω_{ar} but decays with a rate defined by ω_{ai} . Therefore, the decaying rate for each natural mode is,

$$-2\omega_{ai} = \frac{P_{dissipated}}{W_{stored}}. \quad (36)$$

Based on Eq. (36), the “life time” of a natural mode can be defined as $1/(-2\omega_{ai})$. The quality factor Q_{mod} for each natural mode can be defined as,

$$Q_{mod} = \frac{\omega W_{stored}}{P_{dissipated}} = \frac{\omega_{ar}}{-2\omega_{ai}}. \quad (37)$$

When an incident radiation of frequency ω around ω_{ar} is applied, according to Eq. (33), assuming that the α^{th} natural mode is dominant, the response is

$$\{j_n(\omega)\} \approx \frac{\{j_n(\omega_\alpha)\}^T \cdot \{e_m(\omega)\}}{(j\omega - j\omega_\alpha)} \cdot \{j_n(\omega_\alpha)\}. \quad (38)$$

By noticing that $\frac{\{j_n(\omega_\alpha)\}^T \cdot \{e_m(\omega)\}}{(j\omega - j\omega_\alpha)}$ is a scalar, the response $\{j_n(\omega)\}$ near the resonance is almost determined by the excited natural mode $\{j_n(\omega_\alpha)\}$. Since a fixed distribution of stored energy W and dissipated power P is associated with each natural mode oscillating at ω_{ar} , the modal quality factor Q_{mod} renders a good estimation of the quality factor Q_{res} at resonance.

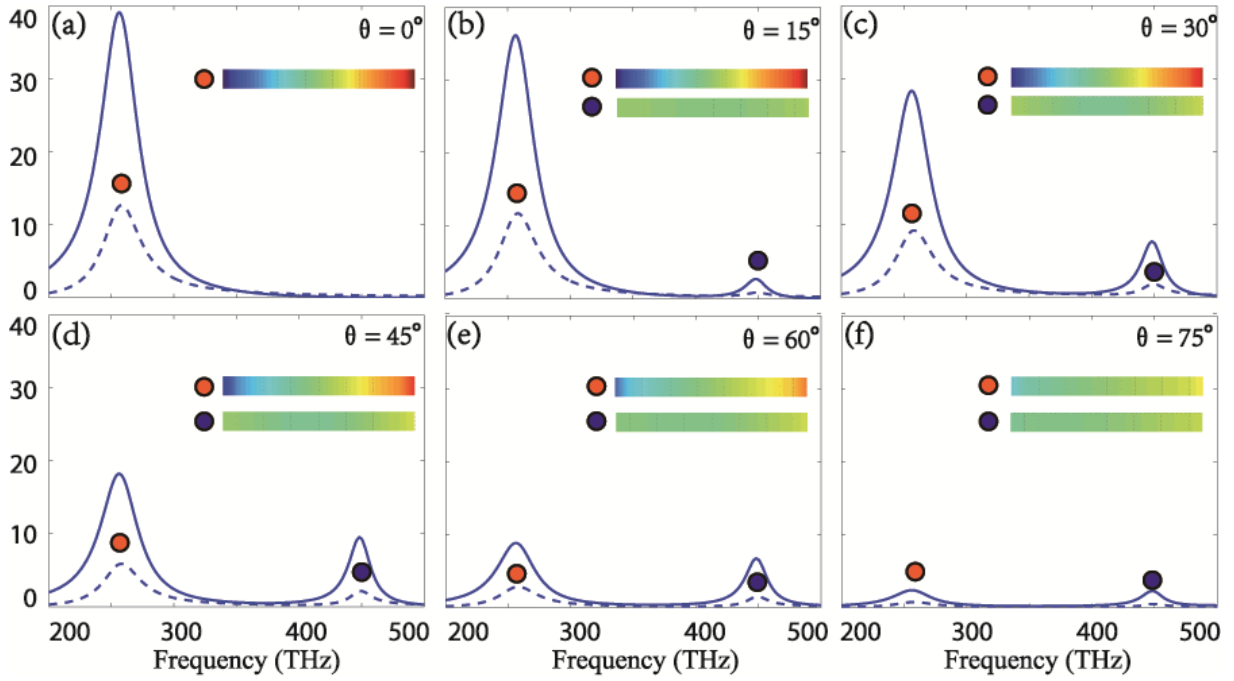


Fig. 15. The stored power (solid line) and dissipated power (dashed line) of the 300 nm nanorod when the nanorod is excited by incident light with various incidence angles. The incident angle and the polarization of the incoming electromagnetic wave are defined in the same way as in Fig. 14. The top surface charge distribution at resonances are shown in the insets and coded from blue to red to denote the negative and positive charge accumulations.

As an illustration, the stored power ωW , the dissipated power P , and the quality factor at resonance (defined as the ratio of the stored power ωW to the dissipated power P at the resonant frequency ω_{res}), when the nanorod of 300 nm is excited by an incident electromagnetic wave with different incidence angles, are calculated from the polarization current flowing inside the structure and plotted in Fig. 15. The details of the calculation can be found in [58]. The quality factor Q_{res} at the resonances and the modal quality factor Q_{mod} are tabulated in Table 3. It can be clearly seen that for the anti-symmetric mode, a good indication of Q_{res} is given by Q_{mod} .

On the other hand, several natural modes can contribute to the formation of one single resonant plasmon mode. In order to illustrate this, the response at the second resonance (450 THz) is calculated by directly solving Eq. (13). The coupling

coefficients $\frac{\{j_n(\omega_\alpha)\}^T \cdot \{e_m(\omega)\}}{(j\omega - j\omega_\alpha)}$ for the anti-symmetric mode and symmetric mode are calculated for different incident angles ($\theta = 15^\circ, 30^\circ, 45^\circ, 60^\circ, 75^\circ$) and shown in Fig. 16. Clearly, not one natural mode contributes to the second resonance. When the incident angle is 15° the contribution from the anti-symmetric mode and the symmetric mode are comparable, and neither the anti-symmetric mode nor the symmetric mode determine Q_{res} , leading to the large deviation from the modal quality factor. As the incidence angle increases, as shown in Fig. 16, the coupling coefficient of the anti-symmetric natural mode decreases. The symmetric natural mode plays a dominant role in the response, resulting in the fact that the quality factor Q_{res} approaches the value of the modal quality factor Q_{mod} of the symmetric natural mode, as shown in Table 3.

θ	f_{res}	$\omega W (10^{-16})$	$P (10^{-16})$	Q_{res}	Q_{mod}
0°	255.8 THz (anti-symmetric mode)	39.09	12.47	3.1336	3.2055
15°		36.13	11.53	3.1336	
30°		28.33	9.053	3.1293	
45°		18.25	5.820	3.1357	
60°		8.843	2.841	3.1126	
75°		2.368	0.7338	3.2270	
15°		2.702	0.8788	3.0746	

30°	450THz	7.474	1.849	4.0422	
45°	(symmetric mode)	9.290	2.151	4.3189	4.6166
60°		6.555	1.473	4.4501	
75°		2.208	0.4725	4.6730	

Table 3. Resonant frequency f_{res} , stored power ωW , dissipated power P , Q_{res} factor at resonances for different incident angle θ and modal Q_{mod} factor of the 300 nm nanorod.

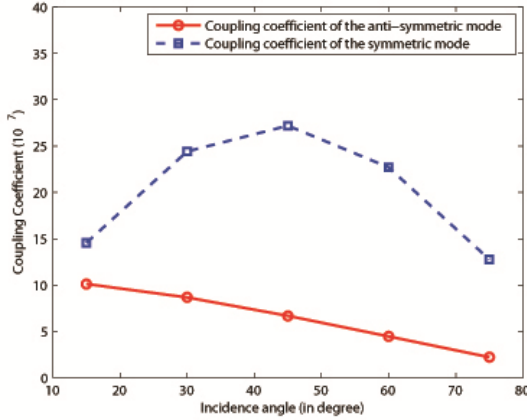


Fig. 16. Coupling coefficient of the anti-symmetric mode and coupling coefficient of the symmetric mode of the 300 nm nanorod.

Having clarified the relation between a natural frequency and the corresponding line position and quality factor of a plasmonic resonance, in this section we apply the proposed full-wave modal analysis to a composite nanoplasmonic system, a Dolmen structure. The Dolmen structure is of great interest in plasmonic research due to its capability of supporting a Fano resonance, known as a sharp asymmetric line shape. This promises applications in nanoscale sensing and the nanoscale analogy of Electromagnetically Induced Transparency (EIT) [59].

The first two natural frequencies and natural modes of the Dolmen nanostructure, made of three 200 nm x 40 nm x 40 nm gold nanorods, are extracted and plotted in Fig. 17 (a) and (c). The gap size between the dimer and the monomer, and the separation distance between the two nanobars in the Dimer are 50 nm and 20 nm, respectively. The whole Dolmen structure is immersed in vacuum and described with 20 nm x 20 nm x 20 nm cells in the numerical study. Again, it can be readily seen from Fig. 17(a) and (b) that the real part of the natural frequencies of the dolmen structure well predicts the line positions of the resonances. Also, it is tabulated in Table 4 that the quality factor of the resonances is determined by the corresponding modal quality factor.

Furthermore, it is interesting to study the physical origin of the line position of the Fano dip in Fig. 17(b) by applying the

full-wave modal analysis technique. The natural frequency of the fundamental natural mode of the individual monomer and the dimer is extracted. For the monomer, the first natural mode is a “superradiant” dipolar mode. Its non-zero dipolar moment radiates into free space, contributing to the main channel for the decay of the stored energy. Therefore, the correspondent natural frequency has a large imaginary part (a short life time and thus a low modal quality factor). As opposed to the monomer, the first natural mode of an individual dimer is an electric quadruple mode whose electric dipolar moment is almost zero, thus only weakly coupled to free space, resulting in a slow decaying and a high modal quality factor. Due to the near degeneracy of these two natural modes, when properly excited, the dolmen structure can be coupled with the incident wave $|I\rangle$ via two channels [60]

$|I\rangle \rightarrow |B\rangle$, which provides a broad background continuum as implied by the low modal quality factor, and $|I\rangle \rightarrow |B\rangle \rightarrow |D\rangle \rightarrow |B\rangle$, where the “bright” mode mediates free space with the high modal quality factor sharp “dark” dimer. The destructive interference between these two channels leads to the Fano dip in the dissipated energy. At the Fano dip, the dolmen structure reacts little to the external electromagnetic perturbation, mimicking EIT at nanoscale.

On the other hand, besides the analysis starting from the interaction between the natural modes of the constituent nanoantennas, the Fano dip can also be understood by closely examining the interaction of the natural mode $|B\rangle + |D\rangle$ with the natural mode $|B\rangle - |D\rangle$ of the whole dolmen structure. First of all, the non-orthogonality of these two modes can be readily read from the surface charge distribution, as shown in Fig. 17(c). This is further proved by numerically calculating the inner product of the two modes. Such a non-orthogonality implies that the excitation of one mode provides a possibility for the other mode to couple with the incident electromagnetic field. At the Fano dip, although the $|B\rangle - |D\rangle$ natural mode still realizes the major contribution, via the near field coupling the $|B\rangle + |D\rangle$ natural mode is excited with the same amplitude but with a π -phase difference. Therefore, a non-active monomer and a non-radiating dimer, as shown in Fig. 17(d), minimize the radiative loss which results in a dip in the dissipated power spectrum.

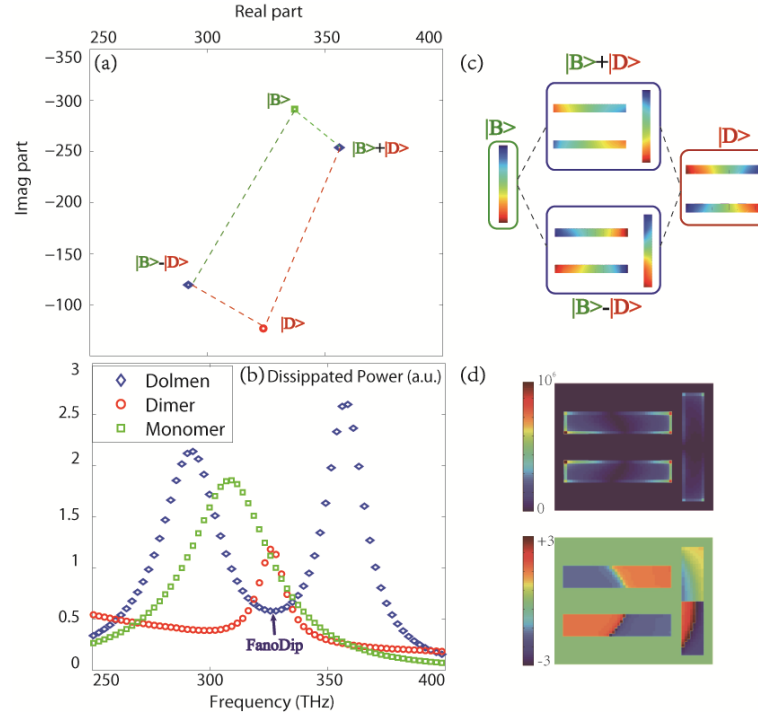


Fig. 17. (a) Hybridization diagram in the complex plane. (b) The dissipated energy of the excited monomer, dimer and dolmen structures. (c) The fundamental natural mode of the monomer and dimer, the first two modes of the dolmen and the propagation direction and the polarization of the incident electromagnetic field exciting these natural modes: $|B\rangle$ is the bright mode for the monomer; $|D\rangle$ is the dark mode for the dimer; $|B\rangle - |D\rangle$ and $|B\rangle + |D\rangle$ are the bonding and anti-bonding mode for the dolmen. (d) The amplitude and phase of the surface charge at the Fano dip.

Structure	f_{real} (THz)	Q_{mod}	f_{res} (THz)	$(\omega W)_{res}$	P_{res}	Q_{res}
Monomer $ B\rangle$	337	3.1026	338.5	2.201e-15	7.245e-16	3.0380
Dimer $ D\rangle$	324	8.0258	324.4	2.671e-17	2.173e-18	12.292
Dolmen $ B\rangle - D\rangle$	292	5.4104	291.5	2.645e-15	4.931e-16	5.3640
Dolmen $ B\rangle + D\rangle$	356	3.6825	357.3	2.071e-15	5.779e-16	3.5837

Table 4. Monomer, Dimer, Dolmen: the real part of the natural frequency, the modal quality factor, the stored power at resonance $(\omega W)_{res}$, the dissipated power P_{res} , and the quality factor Q_{res} at resonance.

VII. CONCLUSIONS

In this paper, we have systematically implemented a set of analysis tools in the framework of Volumetric Method of Moments (V-MoM) algorithm to tackle the light – nanoantenna interaction problem. Such a set is based on the following three linear operator relations which are Eq. (13), Eq. (15) and Eq. (28) respectively,

$$\begin{aligned}
 Z|\mathbf{J}_{scat}(\mathbf{r}, \omega)\rangle &= |\mathbf{E}_{inc}(\mathbf{r}, \omega)\rangle \\
 &\downarrow \\
 Z|\mathbf{J}_n(\mathbf{r}, \omega)\rangle &= \lambda_n(\omega)|\mathbf{J}_n(\mathbf{r}, \omega)\rangle \\
 &\downarrow \\
 Z|\mathbf{J}_{\omega_\alpha}(\mathbf{r}, \omega_\alpha)\rangle &= 0.
 \end{aligned} \tag{39}$$

The line of reasoning behind Eqs. (39) can be clearly seen. The first line corresponds to the most common problem that can be solved by every commercial computational electromagnetic solver and is widely used to confirm experimental speculations. The corresponding solution, e.g. in a V-MoM algorithm the current flowing in a nanoantenna, is

specific for a given incident field and frequency. Consequently, from a design perspective, the first equation in Eqs. (39) sheds very little insight on the intrinsic properties of a nanoantenna. The associated eigenvalue problem in the second line of Eqs. (39) offers a more inherent picture in the sense that its solutions, i.e. the eigenmodes, are incident field dependent but frequency dependent. As demonstrated in Section V, as long as the properties of a metal are solely a function of frequency, the extracted eigenmode is actually independent from the material that constitutes a nanoantenna. Based on this observation, from an antenna engineering point of view, the geometry and material considerations can be individually treated in the first step of a design and combined in later steps. Last but not the least, the solutions to the last equation in Eqs. (39), i.e. the natural modes and the natural frequencies, are both incident field and frequency independent. This set of tools may facilitate the future engineering of optical/mid-infra nanoantennas and bring the full controllability to the THz and optical frequencies.

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